PB84-167642

Decontamination of Hazardous Waste Substances from Spills and Uncontrolled Waste Sites by Radio Frequency In situ Heating

Rockwell International, Newbury Park, CA

Prepared for

Municipal Environmental Research Lab.
Cincinnati, OH

1984

U.S. Department of Commerce
National Technical Information Service
NTIS

PB84-167642

EPA-600/D-84-077
1984

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by

Harsh Dev
Jack E. Bridges
Guggilam C. Sresty
IIT Research Institute
Chicago, IL 60616

Contract Number 68-03-3014

Project Officer

Hugh E. Masters
Oil and Hazardous Materials Spills Branch
Solid and Hazardous Waste Research Division
Municipal Environmental Research Laboratory (Cincinnati)
Edison, New Jersey 08837

MUNICIPAL ENVIRONMENTAL RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
CINCINNATI, OHIO 45268

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TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA-600/D-84-077	2.
3. RECIPIENT'S ACCESSION NO. PB84-167642	
4. TITLE AND SUBTITLE: Decontamination of Hazardous Waste Substances From Spills and Uncontrolled Waste Sites By Radio Frequency in Situ Heating	5. REPORT DATE 1984
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S): H. Dev, J. Bridges, G. Sresty	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Rockwell International, 2421 West Hillcrest Drive, Newbury Park, CA 91320	10. PROGRAM ELEMENT NO. C B R D 1 A
	11. CONTRACT/GRANT NO. 68-03-3014
12. SPONSORING AGENCY NAME AND ADDRESS: Municipal Environmental Research Laboratory--Cin; OH, Office of Research and Developmentd, U.S. Environmental Protection Agency, Cincinnati, OH 45268	13. TYPE OF REPORT AND PERIOD COVERED Paper
	14. SPONSORING AGENCY CODE EPA/000/14
15. SUPPLEMENTARY NOTES: Project officer: Hugh Masters (201) 321-6678	

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT: RELEASE TO PUBLIC	19. SECURITY CLASS (This Report): UNCLASSIFIED	21. NO. OF PAGES: 32
	20. SECURITY CLASS (This page): UNCLASSIFIED	22. PRICE

EPA Form 2220-1 (9-73)

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Harsh Dev

Jack E. Bridges

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IIT Research Institute

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The radio frequency (RF) heating process can be used to volumetrically heat and thus decontaminate uncontrolled landfills and hazardous substances from spills. After the landfills are heated, decontamination of the organic hazardous substances occurs due to thermal decomposition, vaporization, and distillation assisted with steam in a temperature range of 300° to 400°C in a residence time of 14 days. Heating is achieved by laying a row of horizontal conductors above the ground surface of the landfill and exciting them with an RF generator through a matching network. This method is particularly attractive for uncontrolled landfills since it does not require mining, excavation, drilling, or boring in the contaminated volume.

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1. This project has been supported with Federal funds from the U.S. Environmental Protection Agency. Work was performed by IIT Research Institute as a subcontract under EPA contract No. 68-03-3014 with Rockwell International Corp. Mention of trade names or commercial products does not constitute endorsement or recommendation for use by the U.S. Government.

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INTRODUCTION

In the past century, technological advances leading to advanced processes and products have been accompanied by the generation of unwanted waste material. Convenient but inadequate methods for disposing of these hazardous wastes, often by dumping or storing in landfills, have resulted in widely publicized environmental pollution problems (Murray, 1979; Barnhart, 1979; Maugh, 1979, 1979a; Chen, 1978). Accidental spills during transport of hazardous materials have also contributed to the pollution problem.

Hazardous waste materials have been improperly deposited in several thousand sites all over the United States (Maugh, 1979). Some of these wastes, for instance the polychlorinated biphenyls (PCBs), are very stable and can have serious detrimental effects on mankind and the environment. The U.S. Environmental Protection Agency (EPA) has attempted to correct the situation by initiating various regulations that require better management of hazardous waste disposal.

Complete isolation or reclamation of these sites is preferred, but the cost and risk associated with site disturbance by available methods of reclamation are considered prohibitive. New treatment methods are needed that are able to reduce the concentrations of hazardous substances to a level where they are harmless. These methods should be cost effective and not give rise

to problems in other areas by transferring the substances from one biosphere to another.

The radio frequency (RF) in situ heating process has been demonstrated to heat earth and mineral formations rapidly to a temperature of 200°-600°C. The hazardous substances present in a landfill can possibly be rendered harmless by the application of the same in situ heating technology at high temperatures (300°-400°C) following relatively long soaking times (up to two weeks). The process has the following potential advantages for landfill decontamination:

- in situ treatment of most hazardous substances
- no need for excavation, mining, earth-moving, or drilling activities on or through the contaminated volume
- reduced exposure of operating personnel to hazardous substances as opposed to other methods of cleanup
- mobility of major capital equipment
- minimal environmental impact
- small amount of contaminated storage generated requiring special handling.

RADIO FREQUENCY HEATING TECHNOLOGY

The term RF generally refers to frequencies used in wireless communication. The frequencies can be as low as 45 Hertz (Hz) or extend well above 10 gigaHertz (GHz). The frequencies of principal interest to heat earth resources are between 2 and 45 megaHertz (MHz). Proper selection of frequency within this range will ensure electromagnetic wave penetration of a few to 10 meters into typical soil, while at the same time generating average absorbed power densities of about 0.3 watts per kilogram (W/kg) for conservative values of electric field density. The interrelationship between soil

properties and selection of operating frequency is described elsewhere (Von Hippel, 1954; Bridges, et al., 1980).

Earlier dielectric heating designs

Several types of electrical power input arrays have been designed and tested for in situ heating of earth and mineral formations (Bridges, 1980). Most of these systems were designed for thermal resource recovery from deposits of oil shale, tar sand, heavy oil, etc. Invariably, all of the previous designs of RF energy input arrays consisted of electrodes, antennas, or microwave exciters placed in boreholes drilled through the deposit.

In principle, the previous dielectric heating methods are suitable for true in situ volumetric heating of contaminated landfills, but require drilling of boreholes through the contaminated volume. This creates the associated risk of contaminant redispersion, personnel exposure, and fire or explosions from sparks during drilling activities. It is therefore essential to develop an RF energy applicator that is nonradiating and does not require drilling through the contaminated soil in order to heat uncontrolled landfills successfully.

Previous applications of the RF process

The radio frequency heating process has been under development since the mid-1970s for the recovery of hydrocarbons by heating large volumes of earth in situ. After the initial laboratory experiments and the development of computer models to predict the heating patterns based on deposit properties, field experiments were conducted in Utah. The first field experiment was conducted at Avintaquin Canyon, where approximately one ton of oil shale was heated to about 385°C (Carlson, 1980). This was followed by two field experiments at the Asphalt Ridge deposit of Utah. Approximately 30 tons of tar sand were heated using RF energy and about 8 barrels of bitumen were recovered (30

to 38% of the total in place). These field experiments demonstrated that large volumes of earth can be heated volumetrically to temperatures of up to 400°C (Krstansky, 1982). Background levels of the RF radiation in the vicinity of the field test were monitored during the test. It was found that leakage radiation levels did not exceed the recommended ANSI Standard C-95.

IN SITU DECONTAMINATION WITH RF HEATING

The concept of in situ decontamination has three requirements:

(1) a high temperature (300°-400°C) coupled with long residence times (1-2 weeks)

(2) the presence of decontamination mechanism(s) (e.g., thermal decomposition, distillation, vaporization, fixation to soil constituents) for the destruction or mobilization of the contaminants

(3) the presence of a gas and vapor recovery mechanism allowing their collection at the surface.

Decontamination mechanisms

The ability of RF heating methods to heat large volumes of soils and earth formations in situ rapidly can be applied to satisfy the first requirement for the decontamination of soils containing hazardous chemical wastes.

Thermal decomposition. Temperature and residence time requirements for the thermal decomposition of chlorinated hydrocarbons (HCs) were estimated by extrapolating data obtained by Duvall (1980) for the incineration of hexachlorobenzene (HCB). This approach was used to obtain preliminary engineering estimates of the required time and temperature in the absence of data for thermal decomposition at low temperatures and long residence times. Duvall's data show that HCB can be 99.998% decomposed at 1000°C with a residence time of 2 seconds (sec).

Based on these data, a rate constant of 1/2 sec^{-1}, and an assumed reasonable activation energy of 30 kcal/mole, the Arrenhius equation was used to estimate time and temperature requirements. At 344°C, the calculated residence time is 7 days for 99.998% decomposition of HCB. *In situ* treatment can easily provide such large residence times economically because a costly large-volume reactor is not required. Other decontamination mechanisms such as vaporization, distillation, or steam distillation also help in the recovery of contaminants. These can be collected at the surface of the landfill by an appropriately designed vapor barrier and gas collection system.

Distillation. Many compounds found on the CERCLA Hazardous Substance list are hydrocarbons (HC) boiling between 80° and 420°C. Heating these compounds to 300°-400°C would recover a large fraction of the components by vaporization and distillation. Distillation is further assisted by the presence of moisture in the landfill, since, in the presence of steam, the boiling point of the HC/water mixtures is depressed. This mechanism, however, is effective only for those HCs with vapor pressures of the same order of magnitude as water.

In Table 1, the boiling point reduction for some HCs in the presence of water is compared with their normal boiling points. This table shows that steam distillation can be performed with reasonable quantities of steam, provided the vapor pressure of the component is of the same order of magnitude as that of water.

For components boiling at temperatures more than twice that of water, steam distillation requires unreasonably large quantities of saturated steam. Such compounds, however, can be distilled in the presence of superheated steam. The steam acts as a sweep gas that continuously carries the vapors

away from the surface where boiling is occurring, ensuring a good vaporization rate. The PCB mixtures of the Aroclor family, trichlorophenol, and benzidine are examples of hazardous chemicals that can be distilled with superheated steam.

Preliminary estimates were made for the amount of the PCB, Aroclor 1260, that can be distilled with superheated steam generated in situ. This calculation is based on equilibrium and thermodynamic considerations. The results shown in Table 2 are based on the following assumptions: the landfill contains 5 wt% moisture, 25% of the total moisture present in the landfill gets superheated to between 300° and 400°C, approach to equilibrium is only 25%, and the landfill area is 1 acre, depth is 20 ft.

The results in Table 2 show that, with a fixed amount of available moisture, larger amounts of Aroclor 1260 can be distilled as the temperature is increased. Aroclor 1260 was selected in the above example as a worst case since it has the highest boiling range (385°-420°C) of the PCB family.

Vapor and gas recovery mechanism

The RF power will raise the temperature of the landfill so that the decontamination mechanism begins to operate; the decontamination mechanisms will themselves decompose, pyrolyze, distill, and vaporize the contaminants. The vapor and gas recovery mechanisms will allow the gases to escape preferentially towards the landfill surface, where they can be collected for ultimate disposal.

The proposed RF exciter electrode array will develop a temperature and heating profile characterized by a penetrating phase change boundary at which water boils. Figure 1 is a sketch of the landfill showing a horizontal dashed line representing this boundary. The temperature above this boundary will be higher than 100°C, and the temperature below it less than 100°C. In the

higher temperature region (Region A), the permeability of the soil to flow of gases and vapors will be several fold greater than the native permeability of the landfill. The permeability of the landfill will increase due to vaporization of water and the low boiling HCs present in the pore space.

The development of permeability will be directional because it will be confined to those regions where low boiling liquids have evaporated. Thus the region of high temperature, high permeability overlies the phase change boundary for water. The steam will become superheated as it moves towards the surface through the high temperature, high permeability zone of the landfill. On its way to the surface, the superheated steam will sweep the vapors of the higher boiling components present in Region A. The development of directional permeability in tar sand formations was confirmed in the laboratory and the field by Krstansky and coworkers (1982). Figure 2 illustrates the increase in permeability of a tar sand core above the boiling point of water.

A vapor barrier placed above the heated landfill surface will confine and collect the vapors rising to the surface. These gases and vapors will be treated for ultimate disposal by a combination of incineration and on-site treatment.

Process description

A process flow sheet (Figure 3) was developed to allow cost evaluation of the RF decontamination process. A schematic cross sectional view of the landfill is shown in Figure 4.

Rows of horizontal electrodes are placed a short distance above the surface. A vapor barrier is placed over the electrodes. Collection lines carry the gases and vapors to a mobile treatment plant. The vapor barrier is designed to operate under a slight vacuum to prevent venting of the hazardous gases and vapors to the environment.

The impedance matching network, RF transmitters, and other RF hardware (not shown) are placed outside the vapor barrier. Coaxial cables carry RF power to energize individual electrodes. An induced draft (ID) fan carries the gases through a gas handling system consisting of a gas/liquid separator, condenser, cooler, and another gas/liquid separator. The outlet of the ID fan discharges to a demister.

The uncondensed gases from the demister outlet are incinerated on site in a mobile incinerator of the type designed and built by EPA. The liquid phase from both the separators and the demister is collected in a separator where the water-rich phase is separated from the HC phases. The liquid HC phases are also incinerated on site. The water-rich phase is treated on site to produce process-quality water for captive use in the plant.

Other alternative process designs are possible: for example, on-site incineration of all the gases and vapors without condensation, carbon adsorption instead of incineration of uncondensable gases, or treatment of liquid phases at off-site locations.

Treatment duration

The time required to heat a landfill 1 acre in area and 20 feet (ft) deep was calculated for various levels of net power input to the landfill and various temperatures between 300° and 400°C. The duration of treatment was calculated by adding a soaking time of 14 days to the heat-up time.

Figure 5 illustrates the relationship between treatment duration and temperature. For a net power input of 10 MW, the treatment time varies 60 to 90 days as temperature varies from 300° to 400°C. For net power inputs of 2 and 5 MW, the treatment time increases two- to fivefold. Based on these calculations, a net power level of 10 MW was selected for cost evaluation purposes. An RF power source larger than 10 MW is required to account for power

deposition inefficiencies. These include heat loss from the landfill, overheating, and transmission line losses.

Preliminary calculations on heat loss from the landfill show that 15 to 21% of the energy required to heat the landfill may be lost because of thermal conduction. This depends on the treatment duration and the temperature. An overall deposition efficiency of 65% was assumed.

PROCESS ECONOMICS

The cost of landfill decontamination by in situ heating was developed by separately estimating the capital and operating costs for the process. The cost evaluation was based on the following:

- Landfill area is 4047 m^2 (1 acre), depth is 6.1 m (20 ft).
- Treatment temperature range is 300° to 400°C.
- Volatile matter in the landfill ranges between 5 to 20% by weight; 10% of the total volatile matter is organic.
- Power will be provided by 10 2-MW RF transmitters.
- Other than the RF power source, all other process equipment are duplicated so that an average 2.6 sites per year are treated by the single mobile RF source. This is possible because the RF source is required at a site for only 60 to 90 days. The remaining part of the estimated 9 months at each site is used for installing and taking down of equipment.

Capital cost

The capital cost of the process was estimated by developing the cost of each of the seven subsystems of the process separately (Table 3). Vendor quotations were used for all the major equipment items. Estimates were based

on two options: purchased power and on-site power generation. The capital cost for purchased power is $17 million and for on-site generation, $27.5 million.

Operating cost

The operating costs of RF decontamination were calculated by separately estimating the fixed costs for site preparation, equipment mobilization, installation, tear down and decontamination, and permits and licensing requirements (Table 4). These costs are independent of treatment duration. Variable operating costs such as shift personnel salaries, per-diem payments, electric power, water, fuel, and other consumable supplies were estimated on a daily basis. The variable operating cost per day was multiplied by the treatment duration and added to the fixed operating cost to obtain the total operating cost.

Table 4 shows that the estimated total operating cost varies from $3.2 to $4.2 million, depending upon the treatment temperature and volatile matter content. If power is generated on site, then the operating cost is higher by 45 to 54%.

Total cost of decontamination by the RF process

The total cost of decontamination (Table 5) varies between $4.6 to $5.7 million per site. If power is generated on site, then the total treatment cost is higher by 49 to 55%.

The total decontamination cost was obtained by adding depreciation and interest to the operating cost. A uniform straight line depreciation was assumed, with an average interest rate of 22.0% per year.

Cost of decontamination by the incineration process

In the incineration process all the contaminated soil and drums of hazardous substances are excavated from the landfill and shipped to an Annex 1

incinerator. The drums are separately recovered and the waste liquids are transferred to bulk liquid tankers. The soil is loaded in sealed roll-off boxes for shipment.

Our calculations were made on the assumption that an Annex 1 incinerator was available within 250 to 500 miles of the landfill. The incineration cost was assumed to vary between 6 and 16¢/lb.

The operating cost of treatment by excavation and incineration (Table 6) varies between $8.9 to $25.1 million depending upon the volatile matter content, distance to the incinerator, and the incineration cost. This estimate does not include depreciation and interest on capital cost. The interest and depreciation were calculated in a manner similar to that of the RF process, and are included in Table 7.

The total treatment cost for incineration (Table 7) varies between $9.0 and $25.2 million per site. The single largest cost component in this estimate is the incineration cost, which constitutes 79 to 84% of the operating cost. The capital cost for the incineration process (Table 8) is estimated at $0.83 million.

RESULTS AND CONCLUSIONS

A comparison of the costs of decontaminating a hazardous substance landfill by RF *in situ* heating and by incineration (Table 9) indicates that the RF *in situ* process is 2 to 4 times cheaper than incineration.

The RF process also offers significant safety and environmental advantages:

- *in situ* treatment of the hazardous substances
- safe containment of the hazardous waste
- reduced exposure of operating personnel to hazardous substances

- only a small amount of the contaminated waste tonnage generated may require special handling.

The incineration process has already been demonstrated to decontaminate a wide spectrum of wastes found buried in landfills. However, large-scale incineration of contaminated soils may not be immediately feasible due to the inadequate installed capacity of Annex 1 incinerator.

The technical feasibility of the various proposed decontamination mechanisms proposed for the RF process should be verified in laboratory studies.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance and encouragement provided by Mr. Frank Freestone and Mr. Hugh Masters, U.S. Environmental Protection Agency, OHMS Branch, and Dr. Walter Unterberg, Rockwell International, EMS Center. The work reported herein was submitted to Rockwell International in fulfillment of Contract 68-03-3014 under the sponsorship of the U.S. Environmental Protection Agency.

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Table 7. Treatment Costs for Decontamination of Hazardous Waste Landfill by the Incineration Process

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Table 9. Comparison of Decontamination Costs for the RF and Incineration Processes

Component	Mixture Boiling Temp., °C	Pure Component Boiling Point, °C	lb Steam/ lb Component
Benzene	68.3	80.1	0.092
Toluene	83.9	110.6	0.236
Bromoform	94.3	150.0	0.311
Chlorobenzene	91.0	112.5	0.405
Hexachloroethane	98.7	186.0	1.57

% Aroclor in Landfill Mass	Weight of Aroclor Distilled, lb ($x10^6$)	Mass Steam/ lb Aroclor	Required Temperature, °C
1.1	1.43	1.13	300
5.8	7.7	0.21	350
26.0	35.0	0.04	375

Subsystem Description	Total Quantity Required	Cost, $ x 10^6
10, 2-MW RF transmitters, transmission cables, matching network, and dummy load	1	13.33
Vapor barrier and gas collection system	2	0.87
Vapor, condensate, and gas handling equipment	2	1.29
EPA-ORD mobile incinerator	1	1.4
Safety equipment	2	0.07
Instrumentation system	2	0.24
Walkie-talkie radios and charging stations	4	0.007
Total for purchased power option		**17.21**
Add amount shown for power-generation equipment		10.32
Total for on-site power generation option		**$ 27.53**

Volatile Matter, %	Cost ($ x 10^6)				
	Temperature, °C				
	300	325	350	375	400
20	3.86	3.96	4.03	4.10	4.21
10	3.40	3.51	3.61	3.72	3.82
5	3.15	3.26	3.36	3.47	3.56

Volatile Matter, %	Cost ($ x 10^6)				
	Treatment Temperature, °C				
	300	325	350	375	400
20	5.34	5.44	5.51	5.58	5.69
10	4.88	4.99	5.09	5.20	5.30
5	4.63	4.74	4.84	4.95	5.04

Volatile Matter, %	Cost Per Site at ($ x 10^6)			
	400 km to Incinerator		800 km to Incinerator	
	6¢/lb	16¢/lb	6¢/lb	16¢/lb
20	8.9	20.6	9.6	21.4
10	9.6	23.0	10.4	23.8
5	10.1	24.4	10.8	25.1

Volatile Matter, %	Cost Per Site ($ x 10^6)			
	400 km to Incinerator		800 km to Incinerator	
	6¢/lb	16¢/lb	6¢/lb	16¢/lb
20	9.0	20.7	9.7	21.5
10	9.7	23.1	10.5	23.9
5	10.2	24.5	10.9	25.2

Description	Total Quantity	Cost, $
Bulldozer with a ripper	1	320,000
Front end loader with a backhoe	2	160,000
Roll-off boxes with gasketted-hinged doors	300	153,000
Decontamination showers on a mobile trailer		40,000
Trailer mounted mobile laboratory		100,000
Mobile diesel generator set, 75 kW		28,000
Monitoring equipment		24,000
Walkie-talkie radios	4	7,000
Total cost of capital equipment		832,000

Volatile Matter, %	RF Process* $ x 10^6/Site	RF Process* $/100 lb Material	Incineration Process** $ x 10^6/Site	Incineration Process** $/100 lb Material
20	5.34-5.69	4.97-5.30	9.0-21.5	8.4-20.08
10	4.88-5.30	4.00-4.34	9.7-23.9	7.96-19.61
5	4.63-5.04	3.53-3.84	10.2-25.2	7.79-19.26

* Temperature range: 300°-400°C

**Incineration cost range: 6.0¢/lb-16.0¢/lb

Distance to incinerator: 250-500 miles

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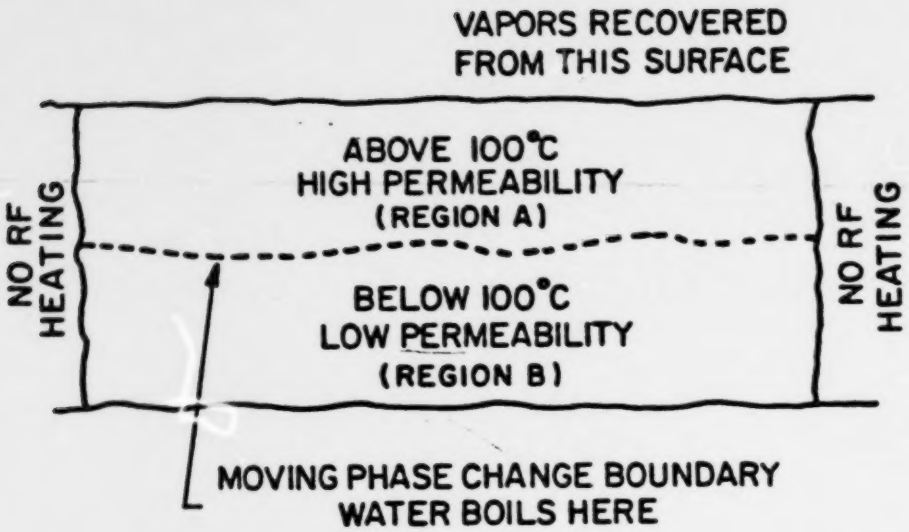
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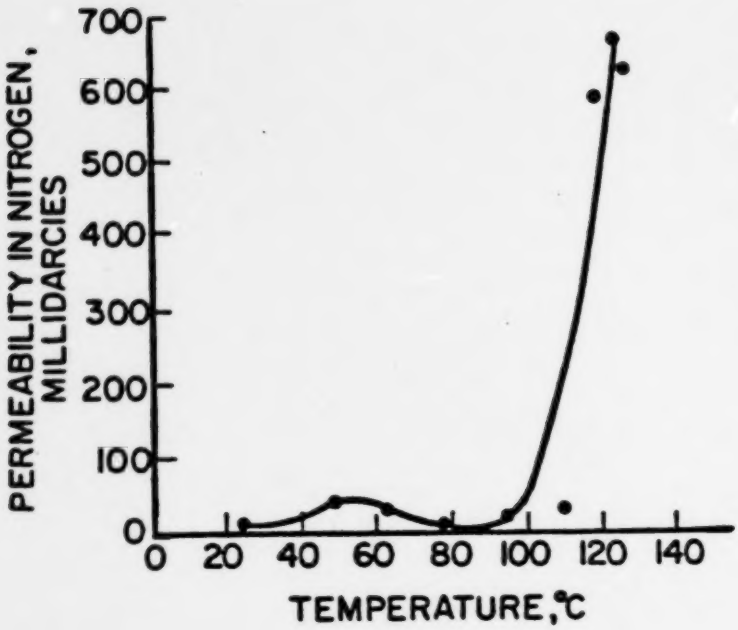
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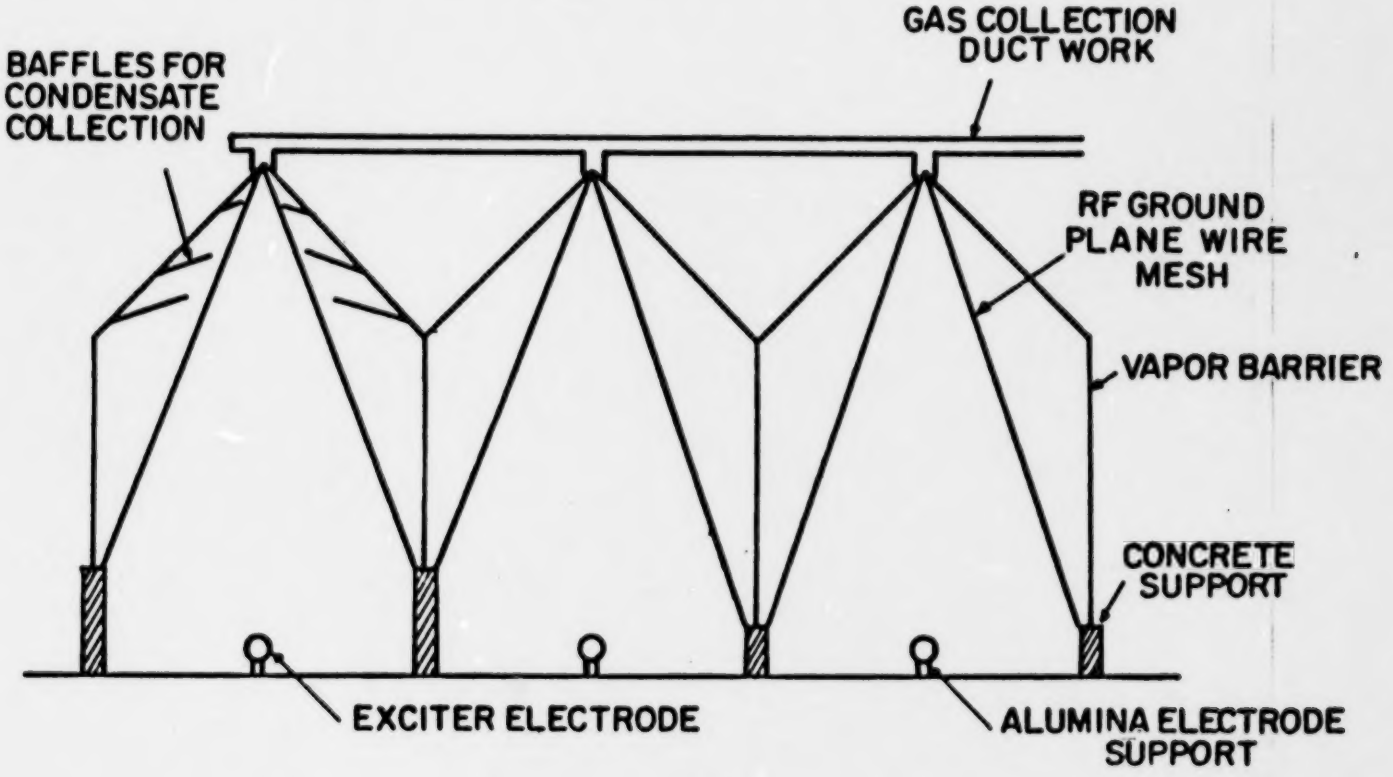
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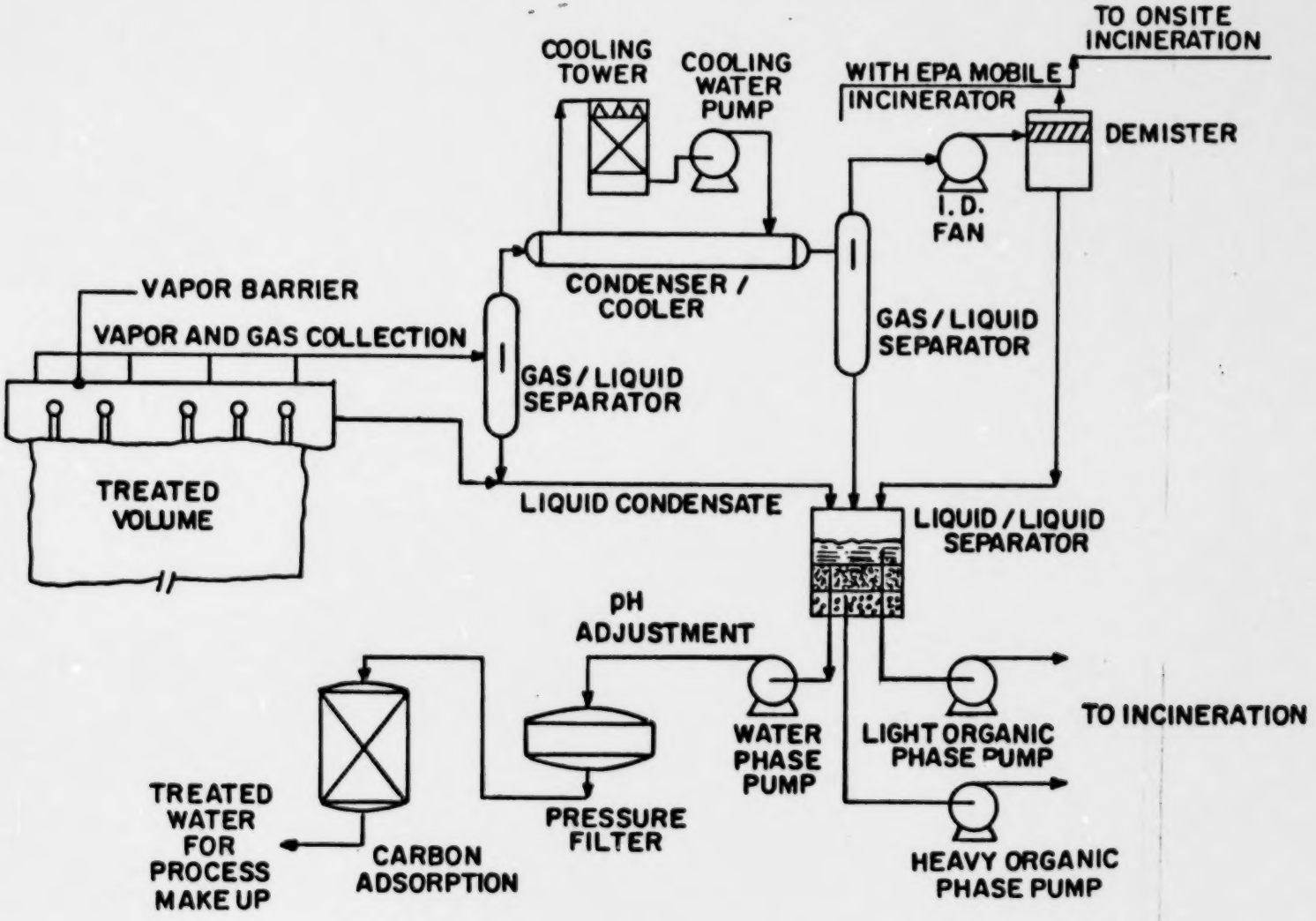
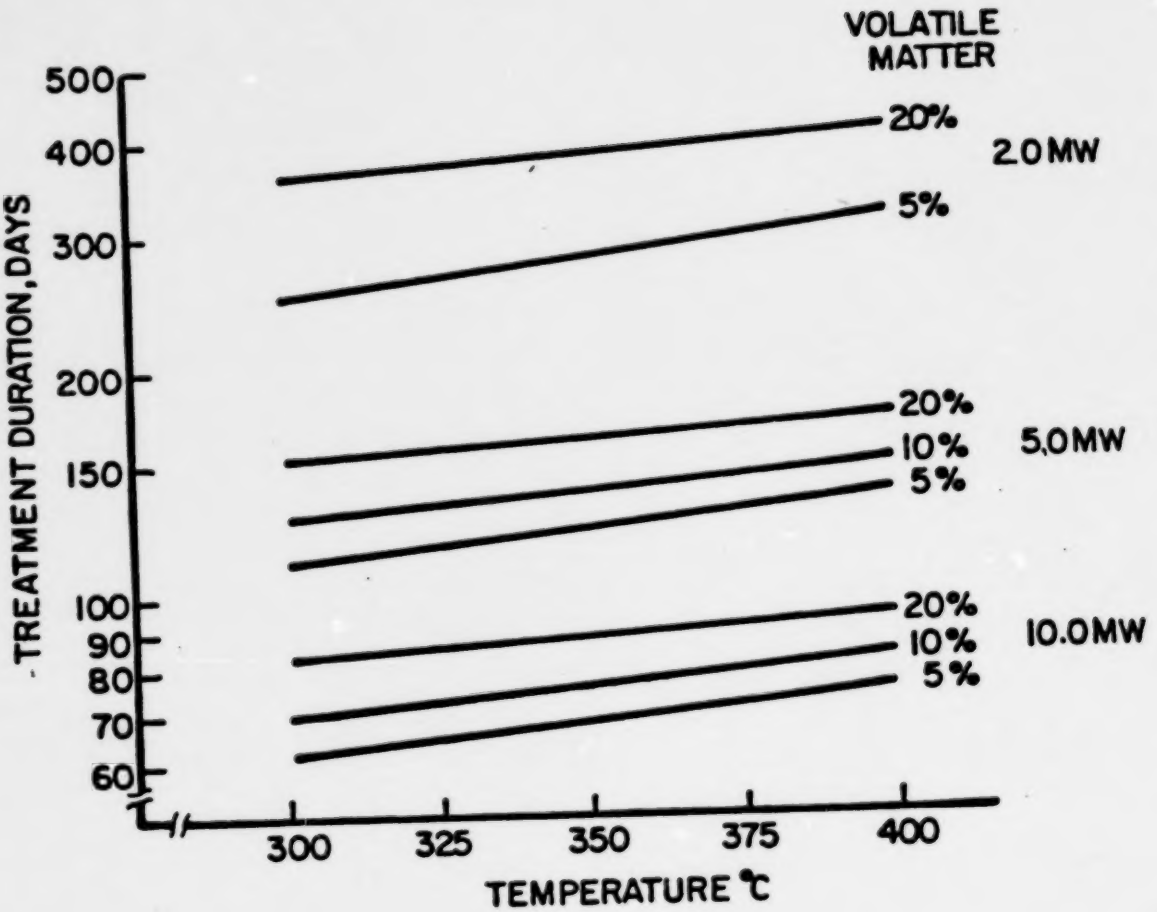
EXCITER ELECTRODE
ALUMINA ELECTRODE SUPPORT


TO ONSITE INCINERATION
WITH EPA MOBILE INCINERATOR
COOLING TOWER
COOLING WATER PUMP
DEMISTER
I. D. FAN
CONDENSER / COOLER
GAS / LIQUID SEPARATOR
VAPOR BARRIER
VAPOR AND GAS COLLECTION
GAS / LIQUID SEPARATOR
TREATED VOLUME
LIQUID CONDENSATE
LIQUID / LIQUID SEPARATOR
pH ADJUSTMENT
WATER PHASE PUMP
LIGHT ORGANIC PHASE PUMP
TO INCINERATION
TREATED WATER FOR PROCESS MAKE UP
CARBON ADSORPTION
PRESSURE FILTER
HEAVY ORGANIC PHASE PUMP


VOLATILE MATTER
500
400
300
200
150
100
90
80
70
60
TREATMENT DURATION, DAYS
20%
5%
2.0MW
20%
10%
5%
5.0MW
20%
10%
5%
10.0MW
300
325
350
375
400
TEMPERATURE °C